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7/25/68

EFFECT OF SURFACE CLEANLINESS ON SOLID PHASE BONDING OF COPPER IN ULTRAHIGH VACUUM

A THESIS

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by

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EFFECT OF SURFACE CLEANLINESS ON SOLID PHASE BONDING OF COPPER IN ULTRAHIGH VACUUM

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I shall always be grateful to my wife for her patience, understanding and encouragement during this period.

The author would like to dedicate this thesis to his parents for their love and continued encouragement in the pursuance of his education.

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SUMMARY

The effect of cleaning the contact surfaces upon the solid phase bonding of copper is studied in ultrahigh vacuum. Cleaning is accomplished through filing in vacuum and it is demonstrated by measuring adhesion of such filed surfaces that reasonably clean surfaces are thereby produced. Comparisons are made between the joint strengths obtained at different bonding temperatures and loads for cleaned and uncleaned surfaces, and for cleaned surfaces which have been purposely contaminated. It is shown that, for bonds made in the temperature range of 25°C to 575°C, stronger joints are obtained with cleaned surfaces at low contact loads. With higher contact loads and bonding temperatures, the effect of cleaning the surfaces on the bond strength is reduced. The effect of heating time is also to reduce the difference in joint strengths obtained with cleaned and contaminated surfaces.

CHAPTER I

INTRODUCTION

In recent years, solid phase bonding has received considerable attention (1). Such interest stems largely from the fact that, by solid phase bonding, materials can be joined without melting any of the parts to be joined (2). This is an important process in joining, metallurgically, dispersion hardened alloys like TD nickel which lose some desirable mechanical properties on heating to temperatures near their melting points (3). This process has also been successfully used to bond refractory metals like columbium, molybdenum, tantalum, tungsten, and their alloys at temperatures less than their recrystallization temperatures (1). With solid phase bonding, it is possible to obtain a joint as strong as the parent metals and also having the same mechanical and physical properties. For this reason, solid phase bonding has been extensively used in the electronics industry for joining kovar, copper, and nickel (4).

Basically, solid phase bonding is the process by which particles or members are joined by atomic forces without the presence of a liquid phase at any stage of the process. If it were possible to bring two perfectly flat and atomically clean surfaces into contact, spontaneous bonding would occur due to interatomic forces. The properties of the bond would depend upon the inherent atomic forces. Under such ideal conditions, diffusion of atoms and/or creep at the interface are not required to achieve bonding. However, real surfaces are neither

perfectly flat nor absolutely clean. Real surfaces consist of large and small asperities which are covered with layers of oxides or other contaminants. To join materials in the solid state, it is necessary to remove the contaminant layers from surfaces and to bring the surfaces into intimate contact. There are several ways of accomplishing this. One is to press the two faying surfaces together with loads exceeding the yield strength of the materials so that gross plastic flow breaks up the contaminant layers and forces the surfaces into intimate contact. This process is commonly known as "pressure or deformation welding." In another process, the parts are pressed together at stresses less than the yield strength and are heated in a protective atmosphere at sufficiently high temperature to disrupt or dissolve the oxide film in a reasonable time. The contact area then increases through diffusional processes. This process is usually known as "diffusion bonding."

It is generally accepted that the important parameters of the solid phase bonding process are pressure, temperature, time, and surface cleanliness. The effects of pressure, temperature and time on solid phase bonding have been extensively studied but very little is known about the effect of cleaning the contacting surfaces on solid phase bonding. The previous studies by Vaidyanath and Milner (5) and Hauser (6) are not conclusive due to the fact that these studies were done at atmospheric pressure or at pressures at which the surfaces become recontaminated within seconds. In these studies, the effect of cleaning the surfaces prior to bonding was nullified to a large extent due to recontamination of the surfaces. Another drawback in these studies is that the extent to which the surfaces were cleaned by the different cleaning

techniques used was not specified. Using an ion bombardment and heating technique to clean the surfaces, Batzer and Bunshah (7) studied the bonding of copper, titanium alloy and beryllium at low temperatures, one tenth to one half of the melting temperature, in an ultrahigh vacuum environment. They observed that welds were not made when the surfaces were not properly cleaned. In their experiments, the cleanliness was not actually measured but was merely judged from severity of ion bombardment conditions. If the results of Batzer and Bunshah for OFHC copper are compared with those of Winslow (8) who did not clean the surfaces prior to bonding, it appears that stronger bonds were obtained with surfaces subjected to ion bombardment and heating. These workers, however, used different bonding conditions and other parameters such as material composition and the initial cold work conditions of the metals were different.

To the author's knowledge, no study has shown in a qualitative, as well as quantitative, way the importance of surface cleanliness in solid phase bonding. The potential of such study lies in that cleaning of the contacting surfaces could allow a reduction in minimum bonding temperature, pressure and time. Reduction of bonding temperature is of great importance in joining materials which lose their mechanical properties if heated to high enough temperatures due to embrittlement, recrystallization and grain growth.

The main objective of the present investigation is to determine the effect of cleaning the contacting surfaces in solid phase bonding. Copper was chosen as the metal to be studied as it can be easily cleaned to a reasonable degree by the simple technique of mechanical abrasion

(9). The present study will be done in an ultrahigh vacuum environment to prevent recontamination of the cleaned surfaces.

CHAPTER II

SURVEY OF THE LITERATURE

At the present time, there is no exact theory that can explain the mechanism of bonding of materials in the solid phase. The reason for the nonavailability of any usable theory is the lack of knowledge about real surfaces, about contact of flat real surfaces, and about the growth of real areas of contact with pressure, temperature, and time of contact. The factors affecting solid phase bonding are numerous and their interaction produces a situation that resists an unequivocal separation of the factor of surface cleanliness.

Although there is no exact theory at the present time that can explain or analyze the effect of cleaning the surfaces on bonding, some insight into this problem can be gained by looking into various phenomena involved and by studying theories or models put forward to explain them.

Model for Solid Phase Bonding Process

A simple, yet very effective, two-stage model for the solid phase bonding process was put forward by Melehan and his coworkers (10). The first stage involves the initial contact of the interfaces. Plastic deformation of the asperities (microscopic roughness) occurs resulting in an initial contact area. The second stage involves growth of the contact area by creep and by diffusion. The diffusion of atoms across the interface, which strengthens the bond, takes place by one of several mechanisms, the most common of which is a jump from hole to hole. The

total area can be expressed as a sum:

$$A = A_o + A_c + A_d \quad (1)$$

where

A_o = initial contact area

A_c = area formed by creep

A_d = area formed by diffusion

Figure 1 shows schematically the bonding process in a single asperity system and the increments of area involved in this model. The macroscopic bonding is visualized as the bonding of a large number of small areas of contact or asperities. All three areas are dependent on temperature. A_c and A_d are also time dependent whereas A_o is primarily load dependent. Based upon this model, the bond strength-time relation at any temperature is expressed as the sum of three components as shown in Figure 2.

This report (10), however, does not take into account the effect of contamination of surfaces on bond strength. Even when asperities deform to support the load, the initial contact area obtained (A_o) may not result in any joint strength if there exists contamination or oxide layer between the contacting areas. This is because the interatomic forces of the metals will come into play only if the surfaces are clean. If, however, due to the load, the asperities deform to the extent that they break the contamination, and thus there is some contact between clean surfaces, the bond will have strength proportional in some way to the contact area of clean surfaces. In the same way, the increase of contact areas by creep and diffusion results in increased bond

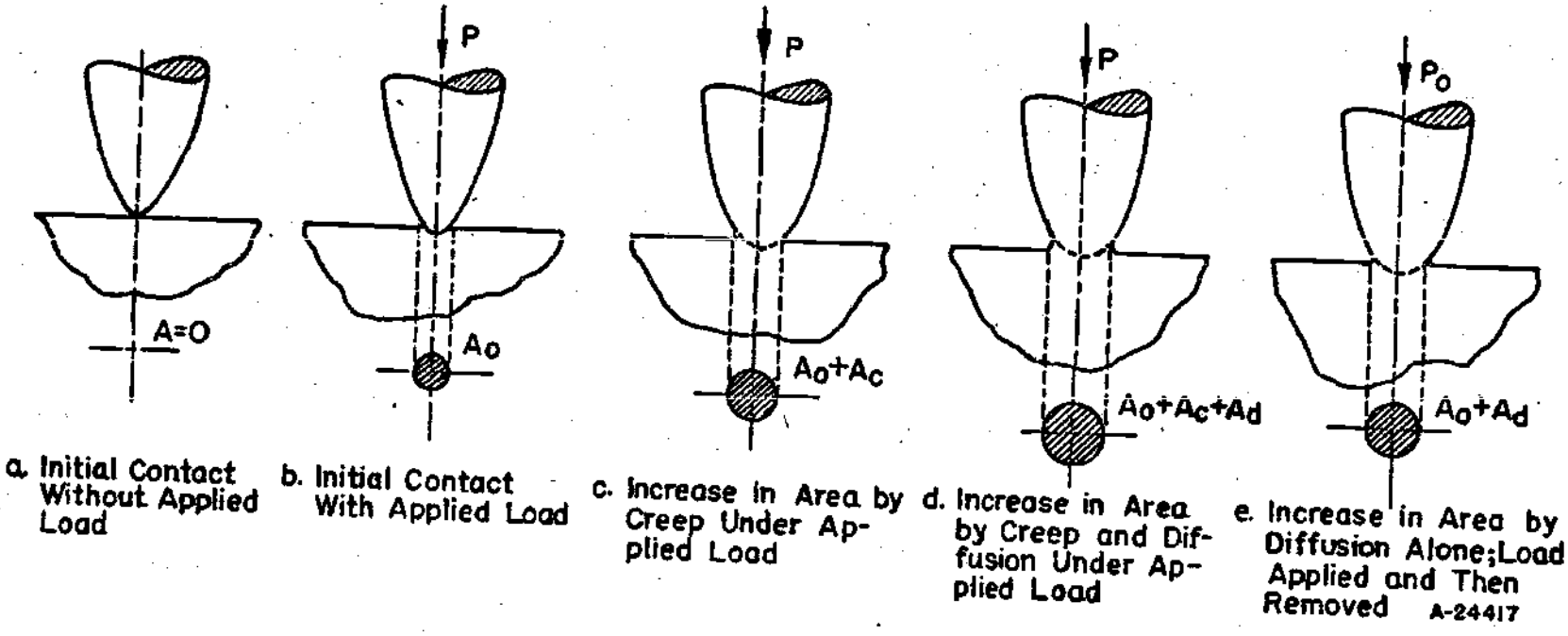


Figure 1. Schematic Diagrams Showing the Effective Bonded Area at Asperity Contact Under Various Conditions of Loading (after Melehan et al. (10))

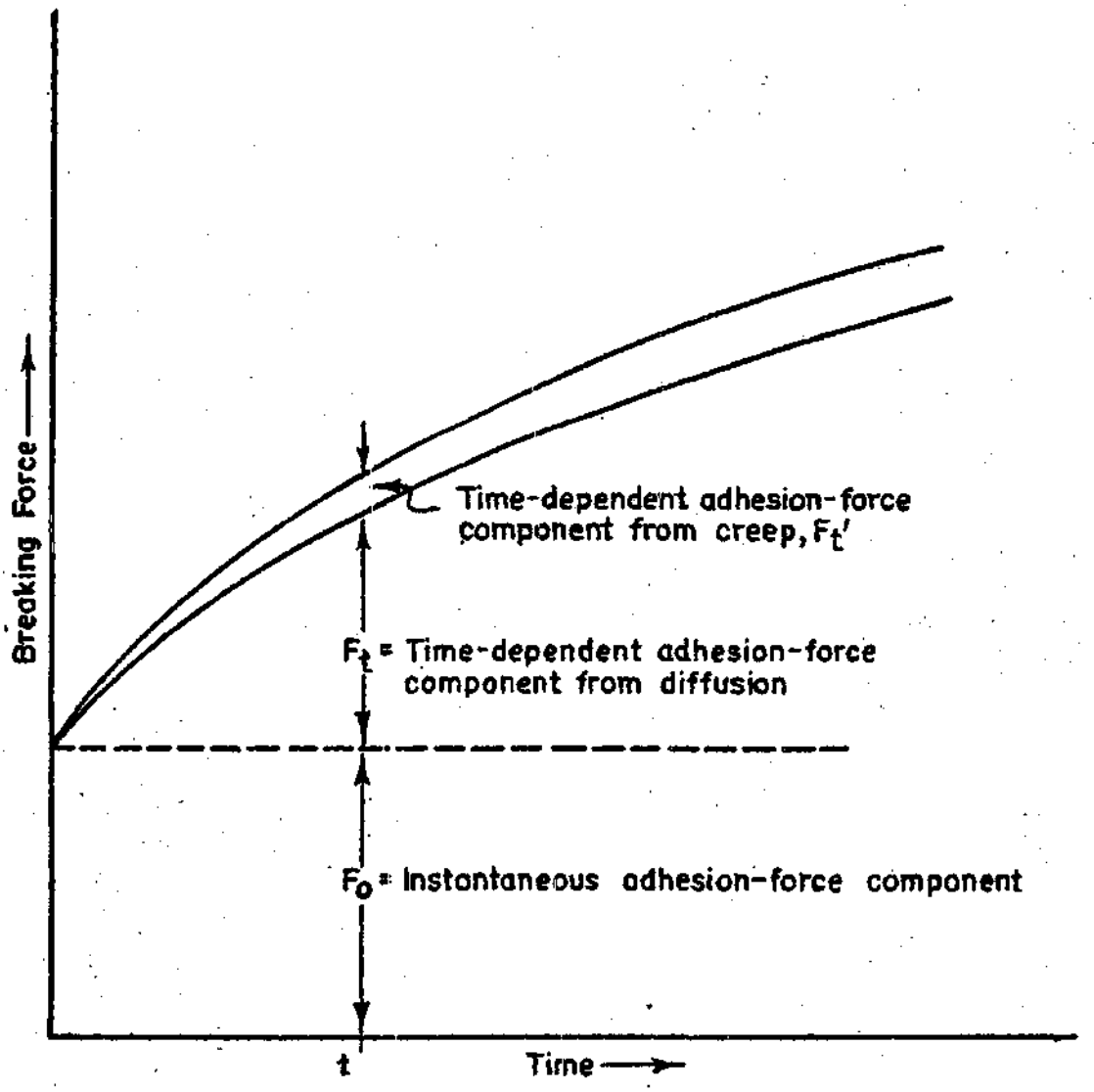


Figure 2. General Form of Breaking Force-Time Curve for Single-Asperity Bonding Tests (after Melehan et al. (10))

strength only if the surfaces which come in contact by these processes are clean.

Using this model, the effect of surface cleanliness on bond strength is the sum of its effects on the three force components (instantaneous adhesion, time-dependent adhesion by diffusion and time-dependent adhesion by creep). Before considering these effects, it is necessary to look into the topology of macroscopic surfaces and the problem of contact of two nominally flat surfaces as this determines the number of points at which surfaces contact and the initial contact area obtained with any load.

Contact Between Rough Surfaces

The Shape of Solid Surfaces

Solid surfaces have been discussed in a number of reviews (11,12). The salient points of these reports are summarized here. Surfaces typically are covered with asperities which are from 10 to 300 micro-inches high and are spaced from 50 to 3000 micro-inches apart. Their slopes occasionally are as steep as 25°, but usually lie between 5° and 10°. The range of summit radii is very large. Many surfaces have height distributions which are Gaussian. Most metallic surfaces exposed to air are covered with an oxide film 10 to 10^5 Å thick (13). The thickness of this film varies with the chemical properties of the metal and with the environment. The oxide film on noble metals may be only several atomic layers thick while on most other metals it is much thicker. In air at atmospheric pressure, a monolayer of gas forms on a surface in about 4×10^{-9} second while at a pressure of 10^{-6} torr approximately three seconds are

required. Layers of adsorbed atoms and molecules are also present on the oxide film.

Real Area of Contact Between Rough Surfaces

Contact between two approaching bodies first occurs where the asperities meet. The asperities resist deformation and cause the real area of contact to be less than the nominal area. This concept of surfaces touching over discrete small areas is fundamental to modern theories of surface contact. The calculation of the area of contact, or even the prediction of how this varies with load, is very difficult. Early attempts to do this by applying the Hertzian theory of contact between spheres to individual contact spots met with the difficulty that the area of the contact spot depends on the radius of the asperity which is normally not known. This obstacle was removed when Holm (14) introduced the idea that although the overall stresses are in the elastic range, the local stresses at the contact spots are much higher so that the elastic limit is exceeded and the contact yields plastically. It automatically follows that the contact area is proportional to load:

$$A = \frac{W}{P_m} \tag{2}$$

where A is the real contact area, W is the load, and P_m is the mean yield pressure determined by the hardness. Objection to this concept was put forward by Archard (15) who showed that for more plausible shapes the area tends to become proportional to load even when contact is elastic.

It was generally thought at that time that the deformation of the asperities was elastic at light loads and became plastic when the load increased beyond some critical value. Greenwood and Williamson (16) have

shown that this is not generally so. In principle, a change from elastic to plastic deformation can occur, but in practice deformation in the contact regions is either always elastic or always plastic. They put forward a theory of elastic contact which combines the elastic behavior of individual asperities with a realistic description of the statistics of the asperity population. Their results show that for a special case of exponential distribution of asperity heights, the number of contact points and the area of contact are exactly proportional to load, that is, the average size of contact points and the contact pressure are independent of the load, and for a Gaussion distribution, these results hold approximately. According to the classical theory of plastic contact, the area should also be proportional to load but experimental evidence (11) does not support this. Recently a new theory for plastic contact of surfaces has been put forward by Pullen and Williamson (17) which takes into account the interaction between individual contact regions. According to this theory, load is not proportional directly to area but is related to it by the relation

$$W^* = \frac{\alpha}{1-\alpha} \tag{3}$$

in which W^* is a dimensionless load defined as $\frac{W}{PA_n}$ where W specifies load, P specifies pressure and A_n specifies nominal area; and α is the degree of contact defined as $\frac{A_r}{A_n}$ where A_r is the real contact area and A_n is the nominal contact area. This relation agrees well with experimental data.

In order to solve analytically the problem of bonding of materials in the solid phase, it is necessary to know the real area of contact for

the macroscopically rough surfaces under different loads and at different temperatures, the number of contact points, the radii of the asperities, the height distribution of asperities, and also how these change when heated surfaces are kept in contact for different times. Present knowledge of surfaces and their contact is limited, as is evident from the discussion above, and it cannot yet supply the information needed to solve quantitatively the problem of solid phase bonding of flat surfaces.

Components of Bond Strength

Instantaneous Adhesion

The tendency for many metallic surfaces to adhere across the interface has been well established under the general term adhesion. Diffusionless adhesion, mentioned as instantaneous adhesion in the model of solid phase bonding, may be attributed to the metallic bonding forces generated by close atomic contact at the interface. As the surfaces approach each other, the so-called long range forces, discussed by Deriagin (18), cause the surfaces to attract one another. When the distance between surfaces becomes less than about 50 Å, the long range forces are reinforced by the short range forces caused by ionic, covalent, dispersion and metallic bonds. The short range forces are generally much stronger than the long range forces (19). The bonding at the interface between the two surfaces is determined by the kind and amount of contamination on each surface as well as by the properties of the surface itself.

Adhesion depends on factors like material properties, load, temperature, surface cleanliness and gaseous environment. It is generally accepted that adhesion is proportional to load; in fact, this has become

known as the second law of adhesion. The validity of this law rests on the supposition that contact area is proportional to load. This supposition, however, has been challenged by a number of workers (11, 17). Detailed discussion of the various factors affecting adhesion can be obtained elsewhere (20, 21). Only the effect of surface cleanliness on adhesion is discussed here.

Effect of Surface Cleanliness on Instantaneous Adhesion

Since the process of adhesion involves the exchange of the free surfaces of two bulk systems (A and B) for a stable interfacial system when the two bulk systems are brought into physical contact, a logical interpretation of the energy balance of the system may be presented by the Dupre equation (22):

$$W_{AB} = \gamma_{A0} + \gamma_{B0} - \gamma_{AB} \qquad (4)$$

where W_{AB} is called the work of adhesion between A and B and γ_{ij} is the interfacial energy (i0 = solid-vacuum and ij = solid-solid). Rabinowicz (23) utilized this equation for the analysis of adhesion and wear phenomena. The Dupre equation predicts that if the surface energy of the bulk phase is decreased, the work of adhesion should also decrease. The surface tension, which is a part of the surface energy, decreases with increasing surface contamination. Thus, the work of adhesion should decrease with increasing surface contamination.

It has been observed by many workers that adhesion of metals decreases with increasing contamination. Gilbreath (20), in his study with atomically clean surfaces, obtained an adhesion coefficient of .78 and .84 for copper and aluminum, respectively. The adhesion coefficient

is defined as the ratio of breaking force to contact force. He observed that contaminating the surfaces by exposing them to an oxygen atmosphere reduced the adhesion coefficient. Approximately 10^{-4} torr-sec exposure to oxygen was found to be sufficient to reduce the adhesion coefficient of copper from .78 to zero. Hordon (9) used wire brushing of sample surfaces in ultra high vacuum to disrupt surface oxide layers. He obtained an adhesion coefficient of .33 at room temperature for copper suggesting that the wire brushing did not remove the contamination fully. With unabraded surfaces, no adhesion was observed. Keller and Johnson (24) and Keller (25) showed in their studies that the normally encountered barriers to adhesion are due solely to surface contamination and that spontaneous adhesion occurs between clean surfaces. They found that the greater difficulty encountered when pressure welding harder metal couples, e.g., molybdenum-molybdenum and couples of hexagonal atomic structure like titaninum-titaninum, is due to the rigorous surface cleaning required for these couples and is not an inherent property of these materials. They concluded that a few atomic layers of contaminants are sufficient to prevent adhesion of these metals under lightly loaded conditions.

Increase of Contact Area and Bond Strength by Diffusion and Creep

As discussed in the two-stage model of solid phase bonding, the bond strength can be divided into three components on the basis of contact area and its growth with time. The first component, instantaneous adhesion, has been discussed previously. The other two components of bond strength, that is, adhesion forces due to diffusion and due to creep, result from the increase of contact area with time by diffusion and creep processes. Here, creep is taken as time-dependent deformation due to

external load only. The growth of the junction for a single asperity system is discussed by Kuczynski (26) for the case of sintering. According to Kuczynski, a growth of interface between a spherical particle and a plane may be represented by an equation of the form:

$$\frac{x^n}{a^m} = Kt \tag{5}$$

where x = neck radius, a = particle radius, K = proportionality constant which includes surface tension, t = time, and

n = 2 m = 1 for viscous or plastic flow

n = 3 m = 1 for evaporation and condensation

n = 5 m = 2 for volume diffusion

n = 7 m = 3 for surface diffusion

Kuczynski, for the purpose of his derivations, defined sintering as the phenomenon of bonding between two or more particles with the application of heat only. His derivations, therefore, have been applied to give growth of a contact junction without the presence of a contact pressure for single asperity adhesion and solid phase bonding experiments.

Holden (27), using the work of Kuczynski, developed an analytic expression for the rate of growth of the bond area in the absence of external pressure (creep) for a single asperity solid state bond. The final expression was as follows:

$$X_o^5\left[(F/F_o)^{\frac{1}{2}} - 1\right]^3 = \left(\frac{81\sigma\delta^3 a^2}{4k}\right)\left(\frac{D_v}{T}\right)t \tag{6}$$

in which X_o = radius of initial contact area, cm; F = breaking force at time t; F_o = breaking force at time zero; σ = surface tension, d/cm;

δ = interatomic distance, cm; a = asperity radius, cm; k = Boltzmann constant, cm/sec; D_v = coeff. of self-diffusion, cm^2/sec; T = temperature, $^\circ K$; t = time, sec. The values for the coefficient of self diffusion determined by using this equation were in reasonable agreement with those given in the literature. As pointed out by Holden, the extension of this equation to a massive system requires more than a simple summation. First, the asperities in a massive system include a wide variation in size, shape and contact pressure. These variables are difficult to evaluate and control. Further, the completion of bonding between massive surfaces requires the elimination of voids through an outward diffusion of vacancies which is not described by the equation. In the literature, there is no treatment of the growth of bonded area when macroscopic flat surfaces are put in contact.

There is still some disagreement about the mechanism of growth of contact area for a single asperity system. Kuczynski (26) concluded from the results of his experiments that at higher temperatures and for large particles volume diffusion was predominantly the mechanism of bonding. Subsequently, Cabrera (28) corrected Kuczynski's calculations with regard to surface diffusion, showing that the theoretical value of the exponent n is 5 for both volume and surface diffusion. He supposed that the different experimental values of n were due to the slight differences in the shape of the metal particles. As discussed by Keller (29), the principal mode of transport is usually considered to be surface diffusion for a single asperity model. Since the effect of contamination films would be to modify the surface diffusion rates, the growth of contact area by diffusion is expected to be dependent on the

nature of the interacting surfaces. The extent of the effect of contaminant film on growth is not treated in the literature.

When the surfaces in contact are under external load, contact area can also increase due to creep. Bowden (30) observed that the contact area of platinum surfaces under static loading at 730°C increased mainly due to creep. Moore and Tabor (31) correlated time-dependent adhesion of indium on steel with creep and stated that the increased adhesion was a direct result of increased contact area. Gilbreath (20), from his experiments with aluminum, copper, lead, and other metals, also thought that increase of adhesion with time was due to creep.

Diffusion of Surface Contamination into Bulk

With contaminated surfaces, diffusion of contaminants away from the interface as suggested by Kinzel (2) and Gilbreath (20) could also lead to increased metal-to-metal contact and higher bond strength. This was not taken into account in the two-stage model by Holden (10). The matter of diffusion of a nonmetallic film, qualitatively, is rather simple. The time-temperature conditions during bonding should be such that the concentration of oxygen at the interface is reduced to well below the maximum value of the solid solubility and is sufficiently small so that reprecipitation on the interface will not occur at lower temperatures due to decreased solubility. Theoretical treatment of the nonmetallic diffusion problem for an idealized case can be found in Kinzel's paper (2).

For the general case of bonding with contaminated surfaces, it

is expected that the above-mentioned time-dependent processes, that is, creep, diffusion and displacement of interfering films in an interrelated fashion, would account for the increase of bond strength with time.

It has been suggested (32, 33) that recrystallization or recovery is the underlying mechanism of adhesion or increase of bond strength. However, it has been observed by a number of workers (1, 27) that recrystallization is not necessarily involved.

Characterization of Surface Cleanliness

Once generated, a surface may be tested for cleanliness by measuring a property of the material which is sensitive to surface contamination. Either physical or chemical properties may be measured. An excellent summary of different approaches that have been used is given in reference (34). A rather simple method of testing cleanliness is by measuring the adhesion between two clean surfaces at room temperature. When two clean surfaces are put in contact with some load, a bond is formed at the interface and some force is required to break the bond. This breaking force is termed as adhesion. As has been discussed, adhesion between metal surfaces strongly depends upon cleanliness of the contacting surfaces, and the existance of one or two monolayers of contaminants at the surface can prevent or reduce the amount of adhesion. In the present investigation, the adhesion at room temperature, in the form of the adhesion coefficient, is used as a measure of cleanliness of contacting surfaces. One advantage of using the adhesion coefficient as a measure of surface cleanliness is that the values of adhesion coefficient for most metal surfaces cleaned by different techniques and/

or contaminated under specified conditions are known. The surface cleanliness obtained by different methods can therefore be compared in terms of the adhesion coefficient. Another advantage is that the adhesion coefficient can be readily measured.

CHAPTER III

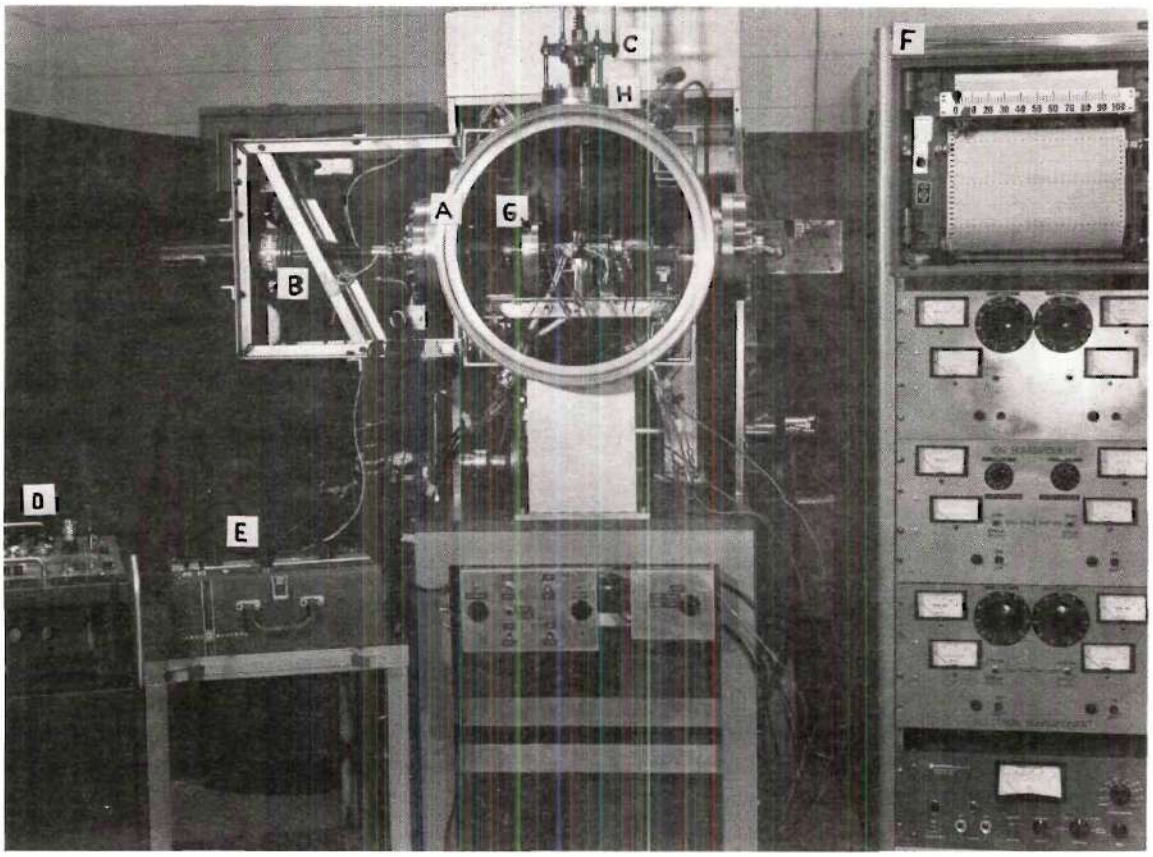
DESCRIPTION OF APPARATUS

Vacuum System

The experiments were conducted in ultra high vacuum to prevent the cleaned samples from becoming recontaminated during the test run. The apparatus was installed in the vacuum chamber shown in Figure 3. The vacuum chamber is a horizontal cylinder 18" in diameter and 39" long. The vacuum system is an oil-free bakeable system pumped by an ion pump in combination with sorption roughing pumps.

In sorption roughing pumps, pumping action is achieved through physical adsorption. At liquid nitrogen temperature, gas is trapped and held in the pump. When the pump returns to room temperature, it is re-activated, that is, the pumped gas is released. A sorption pump removes only a finite amount of gas on each pumpdown. Therefore, the number of sorption pumps required to reduce the pressure to a few millitorr range depends upon the size of the chamber. For the vacuum chamber used, two sorption pumps of 15" overall size were used. The first pump reduces the pressure to 5 torr which is measured on a Bourdon-type gauge and the second reduces it to the millitorr range. The pressure after the second pump was valved in and first valved off was measured on a thermocouple-type gauge.

The 500-liter per second ion pump was used to evacuate the chamber in the vacuum range from 10^{-3} torr to below 10^{-10} torr. The ion pump



A- Vacuum chamber

B- Screw jack

C- File mechanism

D- Sanborn recorder

E- Potentiometer pyrometer

F- Instrumentation for experiment

G- Load cell

H- Chamber flange

Figure 3. Vacuum system and instrumentation

operates on the principle of ion gettering. The pressure in the chamber after it had been evacuated and baked was typically 5×10^{-9} torr. At times, a titanium sublimation pump was used to assist the ion pump in maintaining the low pressure. The vacuum system is essentially the same as is described in reference (35).

Experimental Apparatus

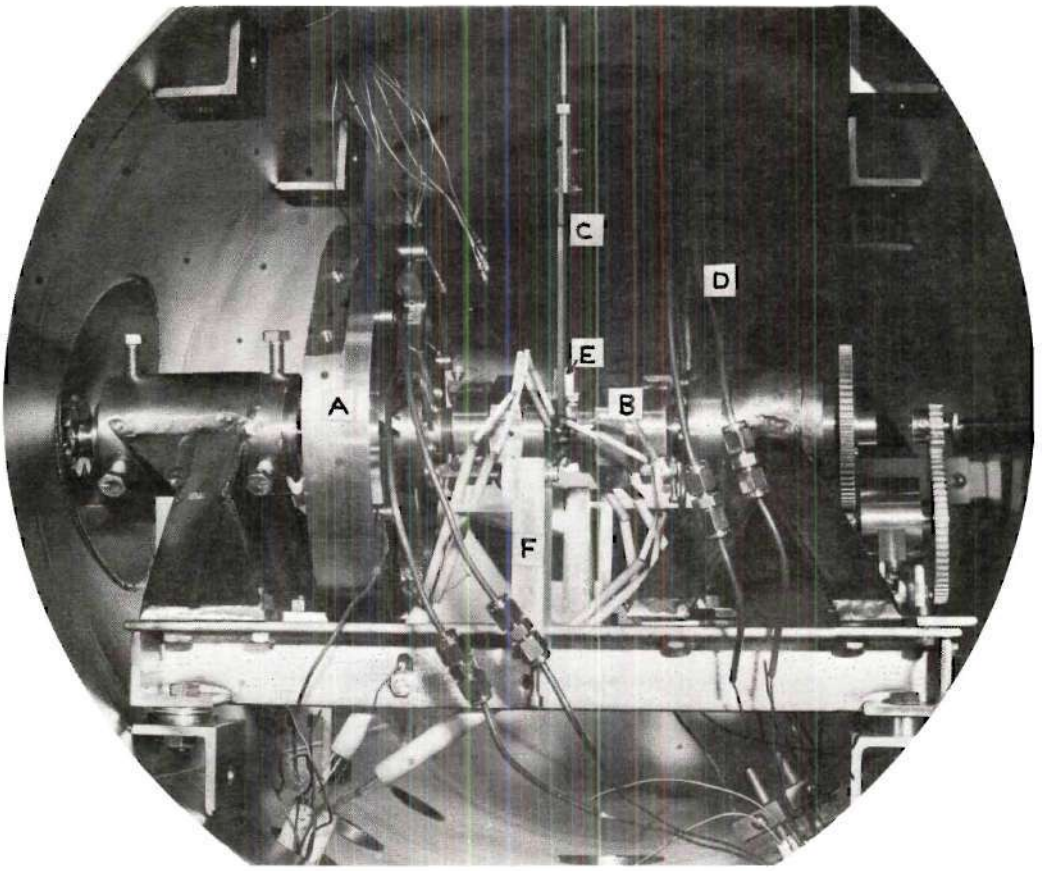
The apparatus installed in the vacuum chamber is shown in Figure 3. The left sample was mounted on the end of a linear motion feed-through and the right sample was mounted in a sample holder bolted to an apparatus which allows axial rotation. During these experiments, however, the right sample was not rotated. A screw jack and variable speed motor were used to translate the linear motion feed-through which moves the left sample. Water-cooled tubes were attached to the sample mounts in order to keep them cool during the specimen heating period and also to reduce the cooling time of the samples.

A close view of the bonding apparatus is shown in Figure 4. The electron gun is, as shown, behind the sample holders. The file is shown hanging above the samples. In order to prevent the moveable left sample holder assembly from sagging during the filing, a support was provided under the sample holder as is shown in Figure 4.

Samples

Samples were machined from high purity copper of the following composition:

Lead	.0015%	Iron	.0002%
Tin	.0004%	Nitrogen	.0002%



A- Load cell

B- Sample holder

C- File

D- Cooling pipes

E- Electron gun

F- Support

Figure 4. Closeup of bonding apparatus.

Arsenic	.0010%	Cobalt	.0001%
Bismuth	.0002%	Manganese	.0001%
Silver	.0004%	Oxygen	.075%
Tellurium	.0002%	Copper	remaining

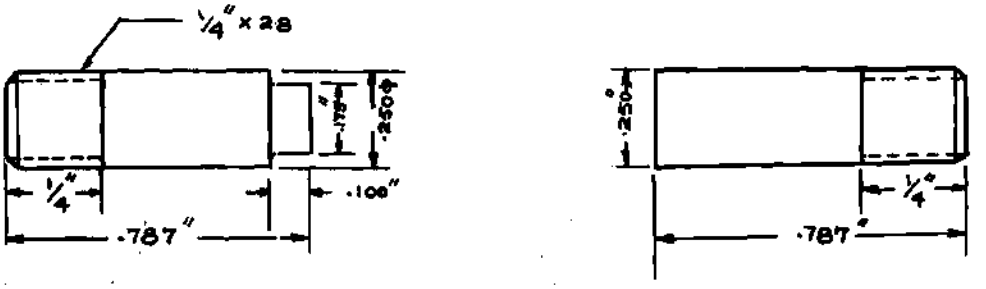
From tensile tests, the yield strength and tensile strength were determined to be 22.6 ksi and 35.3 ksi, respectively. A set of samples with appropriate dimensions are shown in Figure 5. The samples were cleaned with acetone in the ultrasonic cleaner before installing them in the chamber.

Sample Holders

The samples were held in sample holders as shown in Figure 5. The sample holders were designed to be electrically insulated from the samples. Boron nitride spacers between the sample holders and the samples acted as the electrical and thermal insulators. By thermally insulating the samples from the rest of the apparatus, they could be heated to the desired temperature in the order of a few minutes.

Load Cell

The compression force and breaking force of the bond were measured using a specially designed load cell. The load cell configuration resembles a wheel with three spokes extending from the hub to the rim of the wheel. The left sample holder assembly is bolted to the inner hub and the rim is attached to a plate which is, in turn joined to the linear motion feed-through. The plate is keyed to prevent it from rotating. Strain gauges attached to the load cell measure the thrust. The output of this strain gauge type transducer is linear for thrust forces from 0 to 500 pounds, and is measured by a Sanborn recorder.



Left Sample

Right Sample

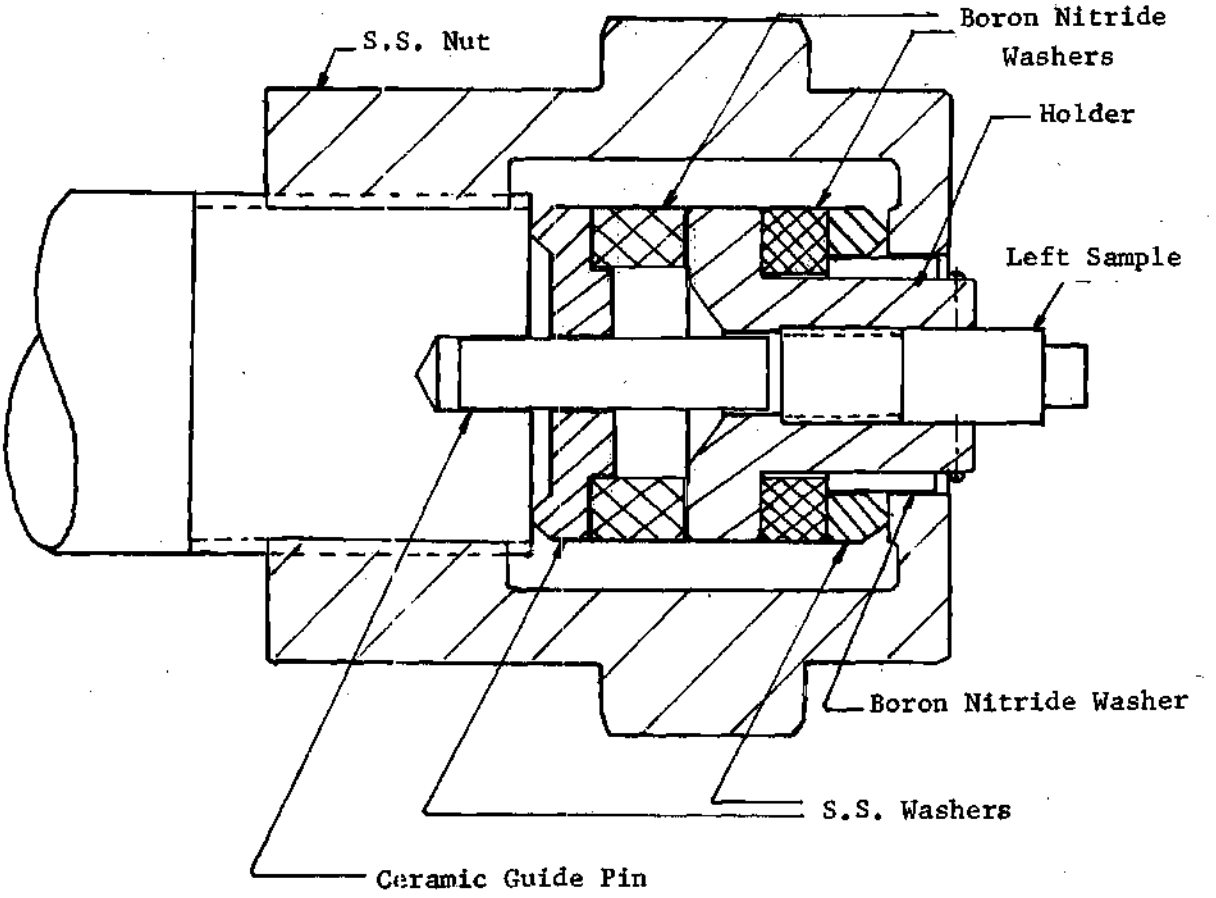


Figure 5. Samples and Left Sample Holder

As previously determined, the transducer had been baked at least ten times to 230°C without any change in the calibration characteristics. There is no noticeable degradation of vacuum level from the outgassing of the load cell. The load cell was calibrated before and after the tests reported on herein with no significant change in sensitivity.

Electron Gun

Figure 6 shows the electron gun used to heat the samples to desired temperatures. The filament is made from .005" diameter thoriated tungsten wire and has about twelve turns per inch. The samples, as well as the filament, were biased during heating and their bias voltages were varied to control the temperature of the samples.

File Mechanism

The file mechanism is shown in Figure 7. The file used was of the Heller 6" single-cut mill smooth type. As shown, the file is attached to the rod which is in turn attached to the bellows feed-through. The movement of the handle up and down compresses and releases the bellows and this in turn moves the file down and up. Before installing the file in the vacuum chamber, it was thoroughly cleaned in the ultrasonic cleaner. To prevent rusting of the file after cleaning, it was installed in the vacuum chamber as the last thing before pumpdown so that it was exposed to the atmosphere for the minimum possible time. After baking the vacuum chamber, no degradation of vacuum was observed due to the outgassing of the file.

Oxygen Inlet System

Commercially pure oxygen was used to recontaminate the samples

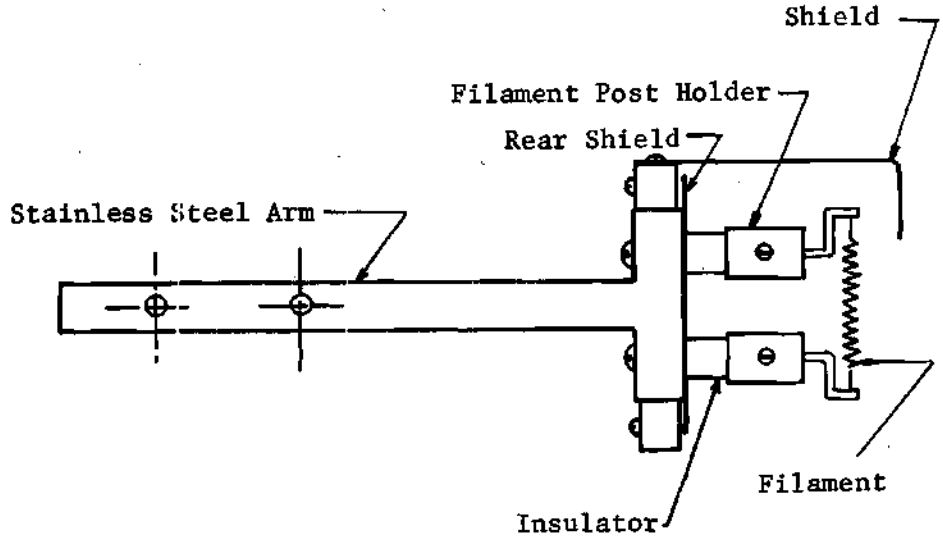


Figure 6. Electron Gun

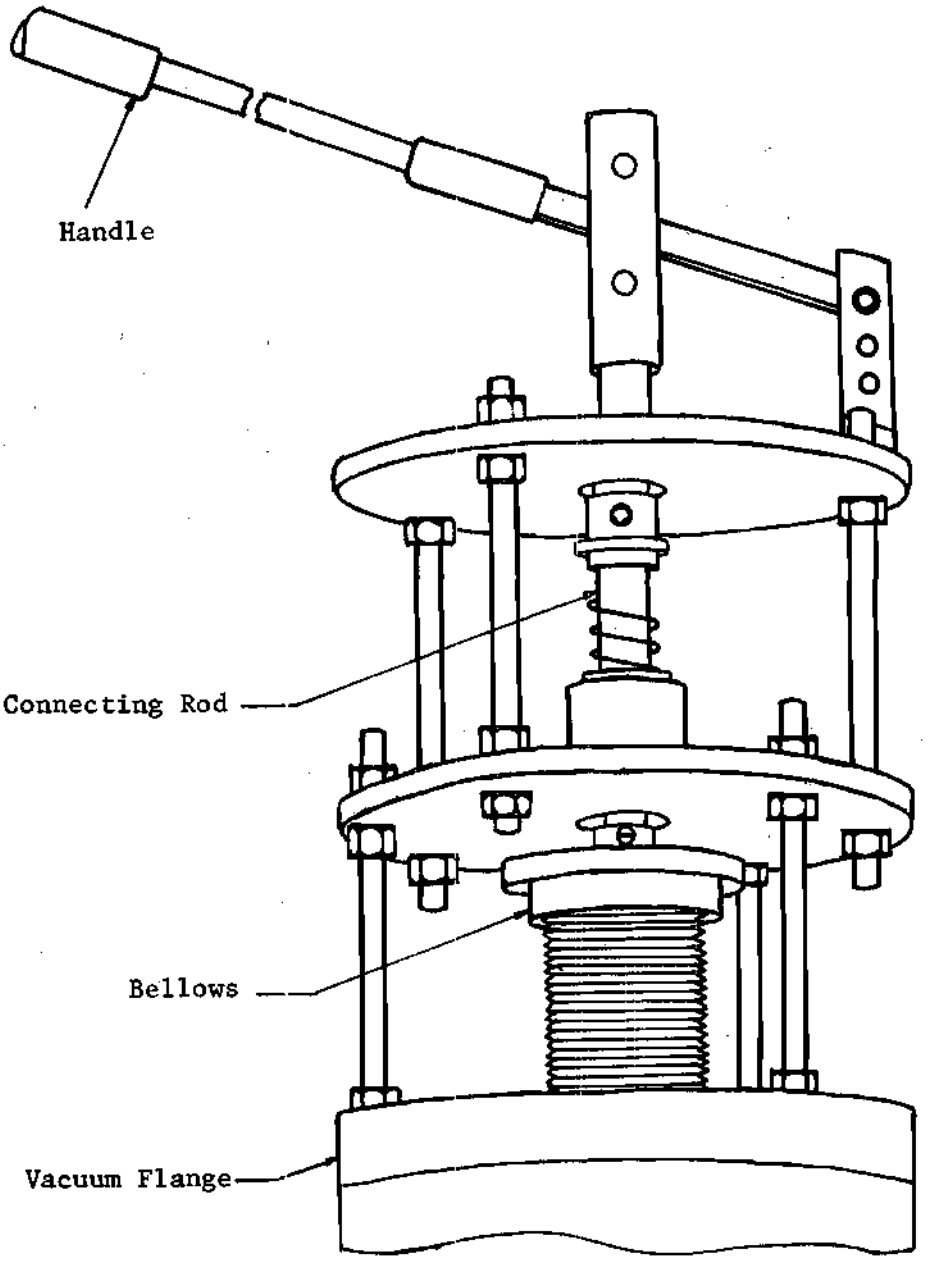


Figure 7. File Mechanism

during certain tests. The oxygen was fed into the vacuum chamber through a variable leak valve. The oxygen cylinder had a pressure regulator to vary the pressure of oxygen flowing to the leak valve.

Temperature Measurement

A chromel-alumel thermocouple was attached to each specimen in order to measure the temperature of the samples. The output of one thermocouple was measured by a potentiometer pyrometer and the output of the other thermocouple was fed into a recorder so that temperature during cooling could be recorded continuously.

Pressure Measurement

The pressure in the vacuum chamber was measured by a nude ionization gauge. The pressure was also indicated by the ion pump control unit.

Electrical Schematic

The electrical schematic for the experimental apparatus is shown in Figure 8. The switches allow heating of the samples and continuous recording of cooling of the samples.

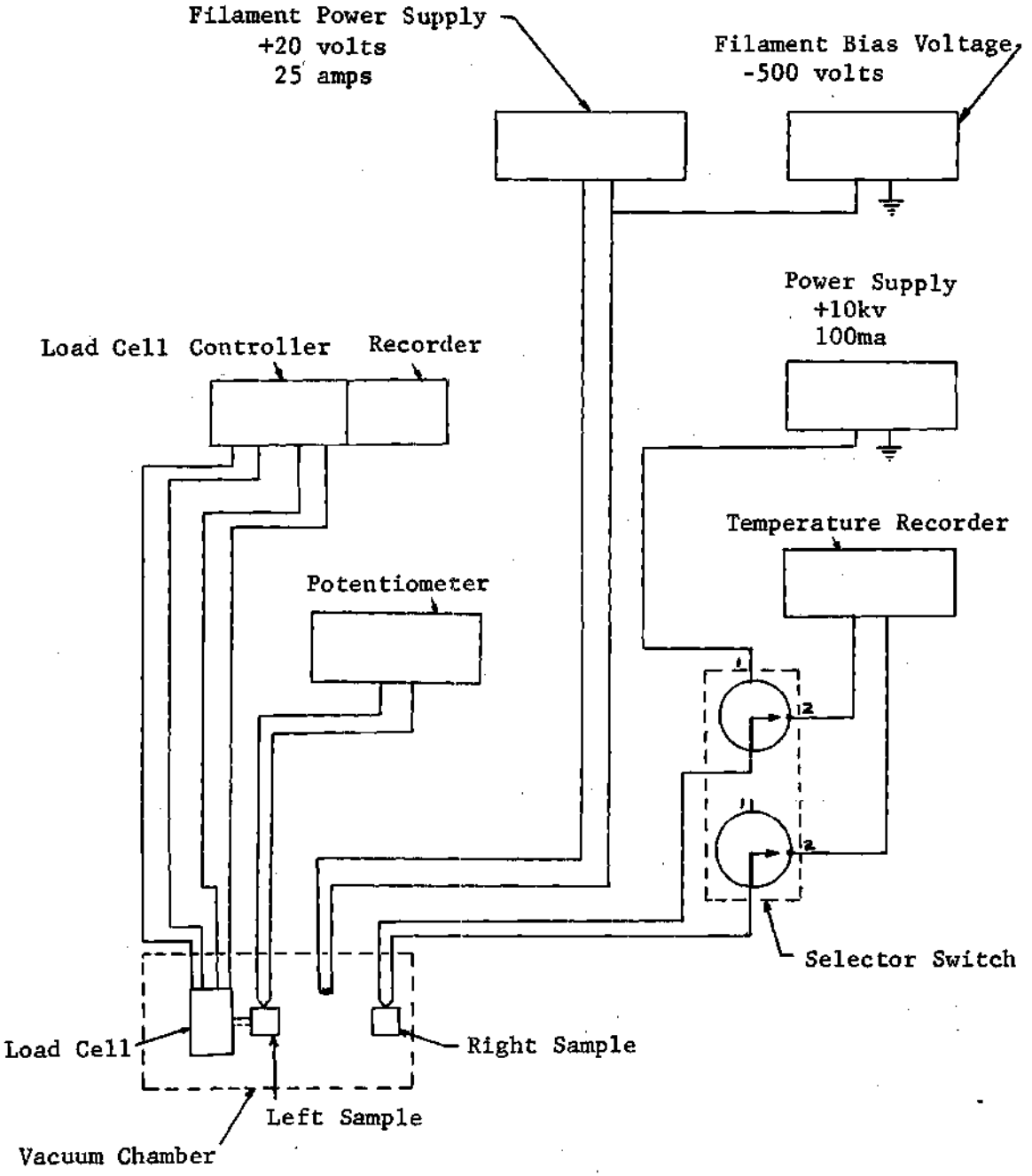


Figure 8. Electrical Circuit

CHAPTER IV

PROCEDURE

After the samples were installed and the connection of thermocouples made, the vacuum chamber was evacuated and baked for approximately 10 hours at 200°C. After the chamber had cooled, the pressure in the chamber was normally in the range 2×10^{-8} to 1×10^{-9} torr. This pressure was considered low enough for the experiments.

The samples were annealed by heating them to approximately 575°C for 30 minutes and then cooling to ambient temperature. The purpose of this annealing cycle was to ensure that subsequent heating at lower temperatures and times did not bring any further metallurgical changes in the structure of the samples. Heating of the samples was accomplished by placing a positive potential on the samples and then electron bombarding them using the electron gun. During heating the samples were put together with negligible load to ensure that any tungsten, evaporated from the filament, did not reach the front faces of the samples. It normally took about 5 minutes to attain a temperature of 575°C.

Cleaning of Sample Surfaces

The procedure for cleaning the samples by filing was as follows. With the samples apart, the file was moved in between the samples. The left sample was advanced inward until the file was held between the samples with a force of 3 to 4 pounds. The file was then moved up and down with the file handle. After a certain number of filing strokes,

normally fifteen, the file was moved out from between the samples. When required, the adhesion coefficient was measured to check the degree of cleanliness.

To determine the adhesion coefficient, the filed samples were pressed together by traversing the left sample inward until the predetermined load (normally 40 pounds) was indicated on the load cell recorder. After keeping this load for a predetermined time, the left sample was traversed outwards at a constant rate until the samples broke apart. The tensile force required to break the samples was recorded on the load cell recorder. The breaking force divided by the compression force gives the adhesion coefficient. Normally an adhesion coefficient greater than .15 at room temperature was obtained with the filed samples.

Measurement of Bond Strength

Bond Strength with Cleaned Surfaces

To determine the bond strength with cleaned surfaces while varying the temperature, the following procedure was used. First of all, samples were cleaned by filing as discussed previously. The cleaned surfaces were then put together with some negligible load (one or two pounds) and heated to the desired temperature by electron bombardment. The samples were then moved apart by moving the screw jack by hand and then immediately put together at the predetermined compressive load by hand movement of the screw jack. The compressive load was maintained for a predetermined time (normally 15 minutes) at the desired temperature. After cooling to 150°F, the samples were broken apart by traversing the left sample outward at a slow contant rate of about 1.8×10^{-3} inches per minute. The tensile

force required to break the samples apart was recorded on the load cell recorder and is called the "bond strength" in this study.

Bond Strength with Contaminated Surfaces

The procedure of determining the bond strength with contaminated surfaces was the same as for finding the bond strength with cleaned surfaces with the difference that an additional step was performed, that of contaminating the filed surfaces with oxygen. The procedure of contaminating the surfaces was as follows. After the samples were filed, they were moved apart as much as they could be to ensure that their faces were fully exposed to the atmosphere of the vacuum chamber. The variable leak valve was slowly opened to let in oxygen. As the ion pump continuously pumped a certain amount of gas out of the vacuum chamber, a steady state pressure in the chamber is attained almost instantaneously for any opening of the leak valve. By adjusting the opening of leak valve, the desired steady state pressure, normally 7×10^{-4} torr (mm Hg), was attained. The surfaces were exposed to oxygen at this pressure for the predetermined time to contaminate them.

CHAPTER V

RESULTS AND DISCUSSION

Discussion of Cleaning by Mechanical Filing

The method of filing samples in vacuum was used to clean the surfaces of the samples. This method is simple and can be easily adapted to vacuum systems. As the surfaces can be reproduced by this technique, this has the additional advantage that many tests can be performed with one set of samples. For studies in ultrahigh vacuum systems where it takes a good deal of time and effort to pump down the system each time, it is an especially suitable method of cleaning the samples. In principle, this method is similar to the other mechanical abrasion techniques like the twist compression technique (35) and scratch brushing (5) which make use of frictional movement to disrupt the oxide film on the surface. These techniques have been used by many workers (5, 36, 37) in pressure bonding and adhesion studies.

To determine the number of filing strokes needed to clean the surfaces of the samples, adhesion coefficients of filed samples were measured after different numbers of strokes. From these tests it was observed that an adhesion coefficient of .15 to .3 at room temperature could be obtained after fifteen strokes and that the adhesion coefficient did not increase with any further increase in number of filing strokes. A similar observation was made by Hordon (9) who cleaned the samples by wire brushing in ultrahigh vacuum. The adhesion coefficient increased

with time of wire brushing until it reached a maximum value of .33 and then no further increase in adhesion coefficient was observed.

Gilbreath (20) used the technique of fracturing notched specimens in ultrahigh vacuum to obtain clean surfaces. With this method, he obtained an adhesion coefficient of .78 for copper which is more than that obtained in this study. The maximum adhesion coefficient obtained in the present study with surfaces filed in vacuum was .3. This difference in adhesion coefficients could be due to any of the following possible reasons. The adhesion coefficient obtained from fracture surfaces, as done by Gilbreath (20), tends to be larger because on a microscopic scale there is some misalignment as the fracture surfaces are put into contact which results in sliding of asperities on one another. This tends to increase the contact area and show larger adhesion values. Another possibility is that surfaces filed in vacuum are not as well cleaned as obtained by Gilbreath by fracturing in vacuum. Yet another possible reason for the difference in adhesion coefficients could be due to the difference in purity of coppers used by Gilbreath and in present study. It has been shown by Sikorski (37) that a very small increase in purity of copper gives a large increase in median adhesion coefficients. Gilbreath used OFHC copper (99.99 percent pure) whereas in this study 99.92 percent pure copper was used. This difference in purity could therefore be the reason for lower adhesion coefficients obtained in this study. The obtained adhesion coefficient of .3 might represent the maximum cleanliness that can be achieved for the 99.92 percent pure copper.

Bond Strength Measurement

Variation of Bond Strength With Bonding Temperature

With Uncleaned Surfaces. Figure 9 shows the results of a test series made to determine the variation in bond strength with bonding temperature for copper samples with uncleaned surfaces. In these tests, the surfaces of the samples were not cleaned by filing prior to bonding. The samples were put together with a compressive load of 40 pounds and heated at the specified temperature for 20 minutes. After cooling to 150°F, the joint was broken in tension to give the bond strength. As is evident from Figure 9, no bond was formed for bonding temperatures less than 200°C. Between 200°C and 400°C, the increase in bond strength with temperature was slow. Above 400°C, the joint strength increased rapidly and uniformly with bonding temperature up to the highest temperature tested. A bond strength of 168 pounds was obtained at 150°F for a bonding temperature of 600°C.

With Cleaned Surfaces. Curve (2) in Figure 10 shows the results obtained in another test series in which the surfaces of the copper samples were cleaned by filing in vacuum prior to bonding. The contact load during heating was 40 pounds and the heating time was 15 minutes. Bond strengths in this test series also were measured at 150°F. As the figure shows, a bond strength of 6 pounds was obtained at a bonding temperature of 25°C. Bond strength increased with increasing temperature up to the highest temperature tested. A joint strength of 215 pounds at 150°F was obtained at the bonding temperature of 575°C.

It may be mentioned here that from the tensile tests, the yield strength (Y_s) of annealed material was found to be 22.5 ksi at room

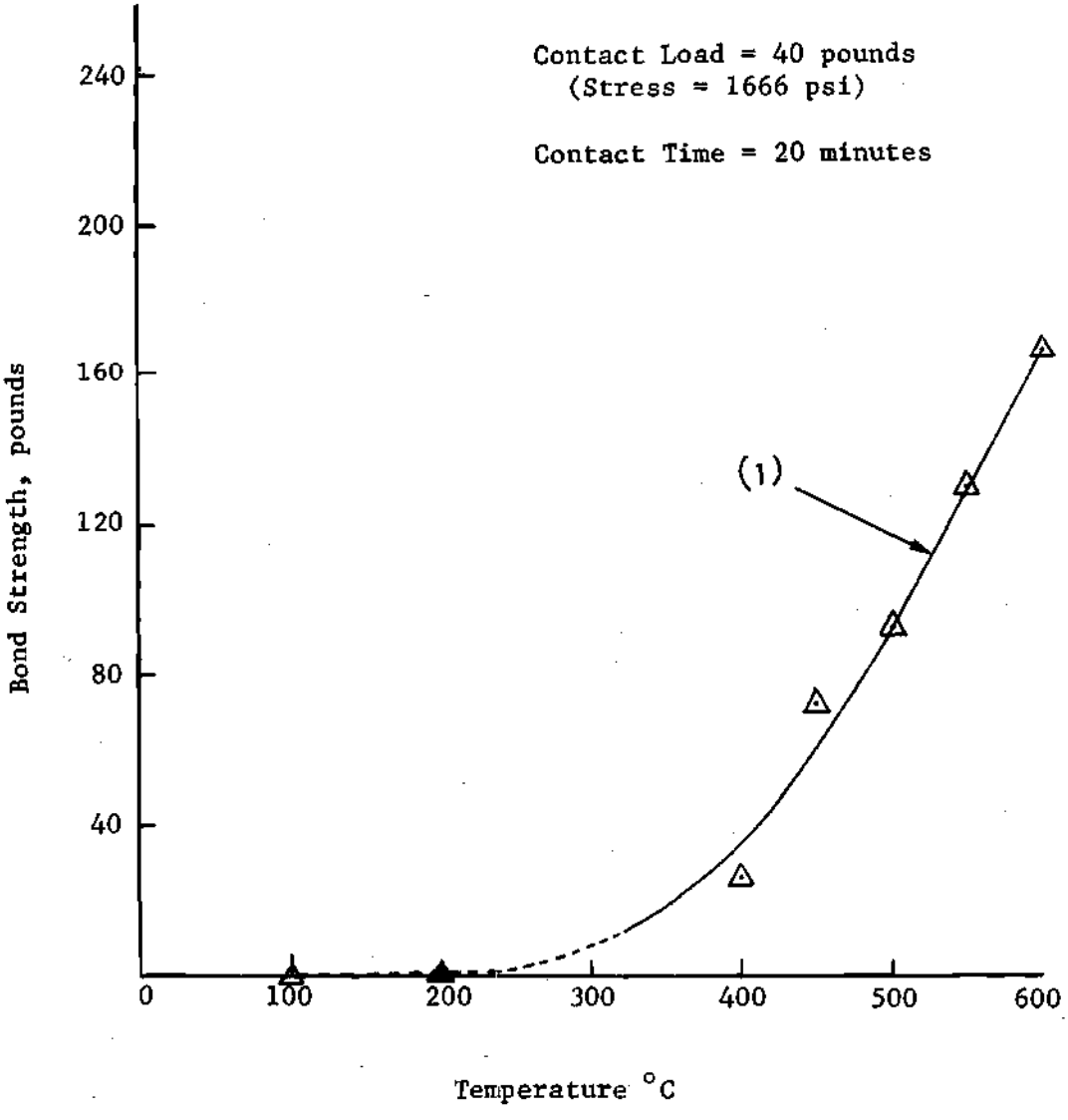


Figure 9. Bond Strength vs. Bonding Temperature With Uncleaned Surfaces

temperature. From reference (38) it seems that yield strength of material would be about 4.5 ksi at 600°C. A load of 40 pounds (1666 psi) is therefore about .075 Y_s at room temperature and about .37 Y_s at 600°C.

Comparison of Bond Strengths. To facilitate comparison, data of joint strength with uncleaned surfaces is also shown in Figure 10. The curves differ essentially in two respects. First, the curve for cleaned surfaces always lies above the curve for uncleaned surfaces implying that the joint strength is always more for clean surfaces for the specified conditions of contact load and heating time. From the curves it is evident that the same joint strength can be obtained at a lower heating temperature by initially cleaning the surfaces of the samples. For instance, a joint strength of 102 pounds at 150°F can be obtained with cleaned surfaces at a bonding temperature of 400°C whereas with no initial cleaning, the bonding temperature must be 520°C to achieve the same bond strength. A reduction in the required bonding temperature can therefore be achieved by initially cleaning the samples. Second, after about 400°C, the slope of the curve for uncleaned surfaces becomes greater than that of the curve for clean surfaces implying that there is a more rapid increase in bond strength with bonding temperature for uncleaned surfaces. Such a behavior can be expected for copper as solubility of oxygen in it (copper) is high and it increases with increasing temperature (39). Therefore for copper, diffusion of surface contamination into the bulk becomes more and more pronounced as the temperature is increased and this tends to reduce the difference in joint strengths obtained with cleaned surfaces and uncleaned surfaces as the bonding temperature is

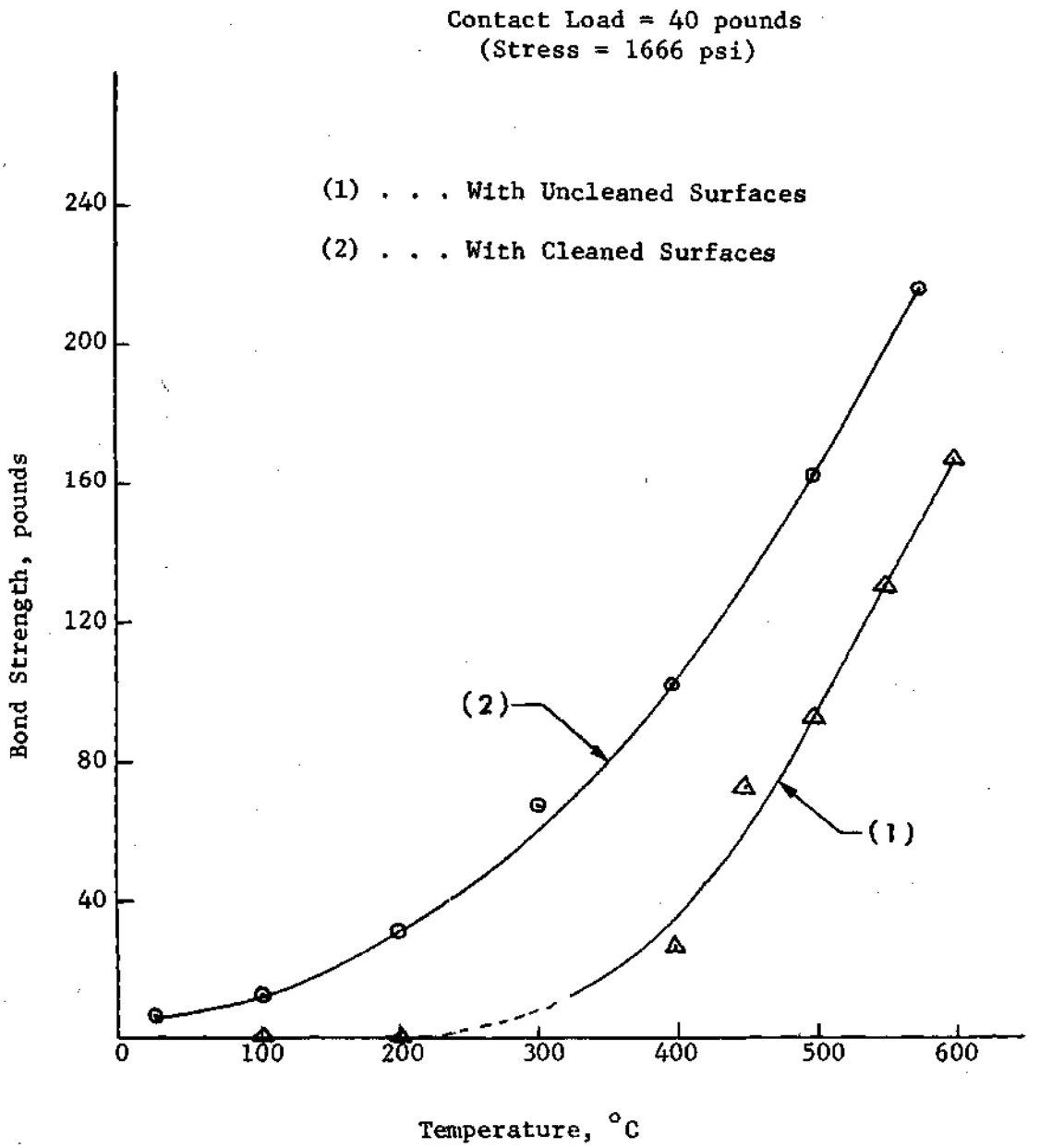


Figure 10. Bond Strength vs. Bonding Temperature for Cleaned Surfaces and Uncleaned Surfaces

increased. The above behavior may not be observed in bonding metals like aluminum for which the oxide will not dissolve into the bulk with increasing temperature due to the negligible solubility of oxygen in them (39).

It may be mentioned here that the curve (1) for uncleaned surfaces was obtained with heating time of 20 minutes whereas curve (2) for cleaned surfaces was obtained with heating time of 15 minutes. This difference in heating time does not affect the comparison of results given above as the bond strength is not a strong function of heating at these times as will be shown later.

It has been suggested (40) that residual cold work locally at the surface as a result of mechanical abrasion may accelerate bonding. This factor could affect the comparison made between bond strengths for cleaned and uncleaned surfaces as cleaned surfaces are additionally subjected to mechanical abrasion. To remove this possible source of error, it was decided to intentionally recontaminate the surfaces after filing them and to use contaminated surfaces in place of uncleaned surfaces in determining the effect of cleaning. Since the contaminated surfaces are also filed, they have the same cold worked condition on the surface as the cleaned surfaces have. The next step then was to determine the extent of contamination obtained with exposing surfaces to different amounts of oxygen.

Change of Bond Strength with Oxygen Exposure

It has been shown by Gilbreath (41) that for copper only the adsorption of such chemically reactive species as oxygen, rather than physically adsorbed gases, affects the strength of joints formed at room

temperature. For joints formed at higher temperatures, physically adsorbed gases are therefore not expected to affect the bond strength.

Figure 11 shows the effect of exposing the cleaned surfaces to oxygen on the strength of bond made at 300°C temperature. In this test series the sample surfaces were cleaned by filing in vacuum and were then exposed to oxygen at room temperature for a specified time and at a specified environmental pressure. Samples were then put together, heated at 300°C for 15 minutes and then broken in tension at 150°F to give the joint strength. After the joint strength had been measured, the surfaces were recleaned by filing and exposed to a higher dosage of oxygen. The results of the test series show that the bond strength was not affected by exposure up to 6×10^{-4} torr-sec of oxygen. With higher exposures, bond strength decreased rapidly until it reached a value of 20 pounds with 3×10^{-2} torr-sec exposure. A further increase of exposure to 8.4×10^{-2} torr-sec, however, did not change the bond strength. In the past, no similar study has been done in the field of solid phase bonding with which the present results can be compared.

In the present study, when the joint was made at room temperature with 2 minutes contact time, the adhesion or strength of the joint decreased to zero for exposures greater than 3×10^{-3} torr-sec. This is consistent with the results of Gilbreath (41) who found that for copper, the adhesion reduces to zero for exposures greater than 10^{-4} torr-sec.

Variation of Bond Strength with Bonding Temperature for Contaminated Surfaces

Curve (3) of Figure 12 presents the results of tests in which the filed surfaces were contaminated with 7×10^{-2} torr-sec exposure to oxygen

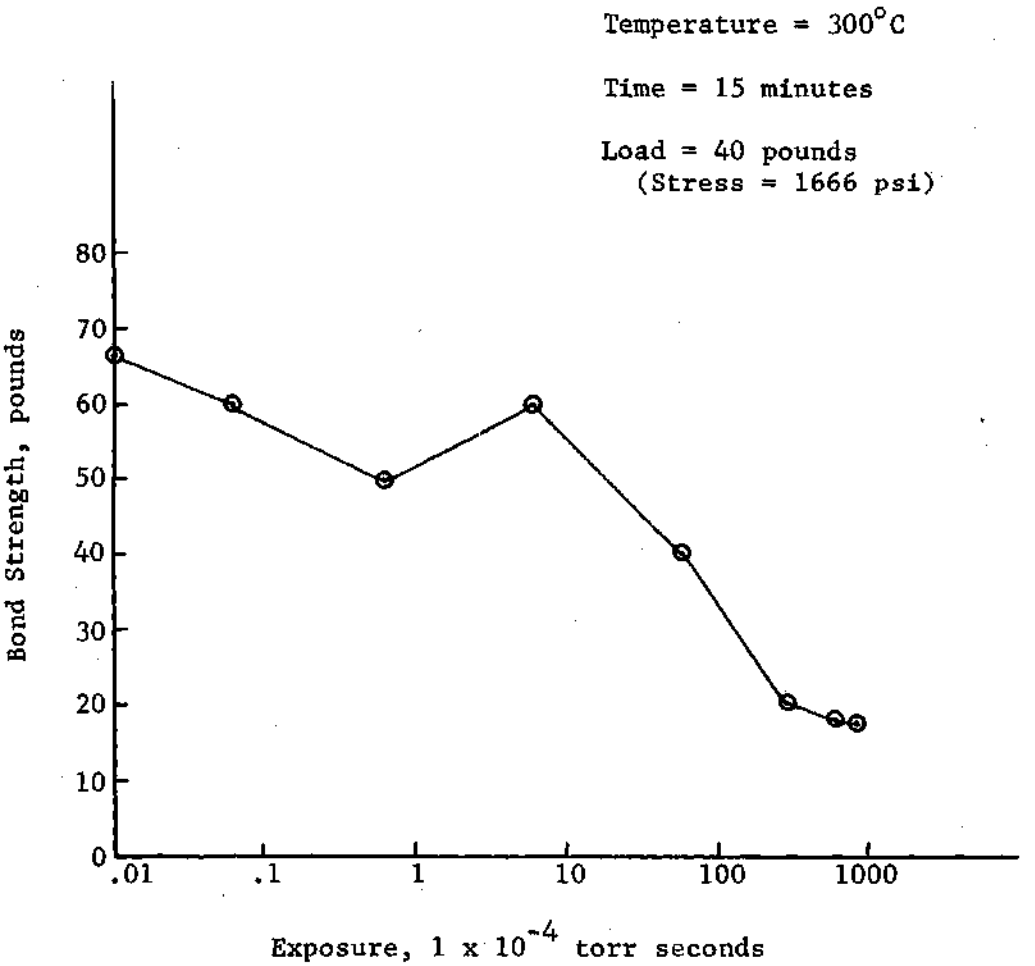


Figure 11. Variation of Bond Strength With Oxygen Exposure

prior to bonding. The contact force was 40 pounds and the heating time was 15 minutes. To facilitate comparison, curves (1) and (2) of Figure 10 are also shown in Figure 12. For curves (2) and (3) it can be readily seen that the effect of contaminating the surfaces is to reduce the bond strength for the specified condition of contact load and time. Comparison of curves (1) and (3) shows that the reduction is not as much as was observed with the uncleaned surfaces. The bond strength at various bonding temperatures with contaminated surfaces is more than that obtained with uncleaned surfaces. Uncleaned surfaces, by virtue of being exposed to air at atmospheric pressure, are contaminated with a much higher order of exposure of oxygen. An oxide layer can therefore be assumed to be existent at the surface. On the other hand, exposure to 7×10^{-2} torr-sec of oxygen can, at the most, form only a thin contaminant film of a few monolayers. It can be easily seen that greater energy will be required to dissolve or disrupt the oxide layer in the case of uncleaned surfaces than to disrupt the thin film in the case of surfaces contaminated with 7×10^{-2} torr-sec exposure to oxygen. It can therefore be expected that for the same contact load, bonding temperature, and time, the bond strength with uncleaned surfaces will be less than the bond strength obtained with contaminated surfaces. It should be noted that the difference in bond strength between the contaminated and uncleaned surfaces is not much which implies that a few monolayers of contaminant film on the surface can effectively inhibit the bonding of surfaces. This points out the advantages of performing the solid phase bonding process in a controlled atmosphere or in vacuum.

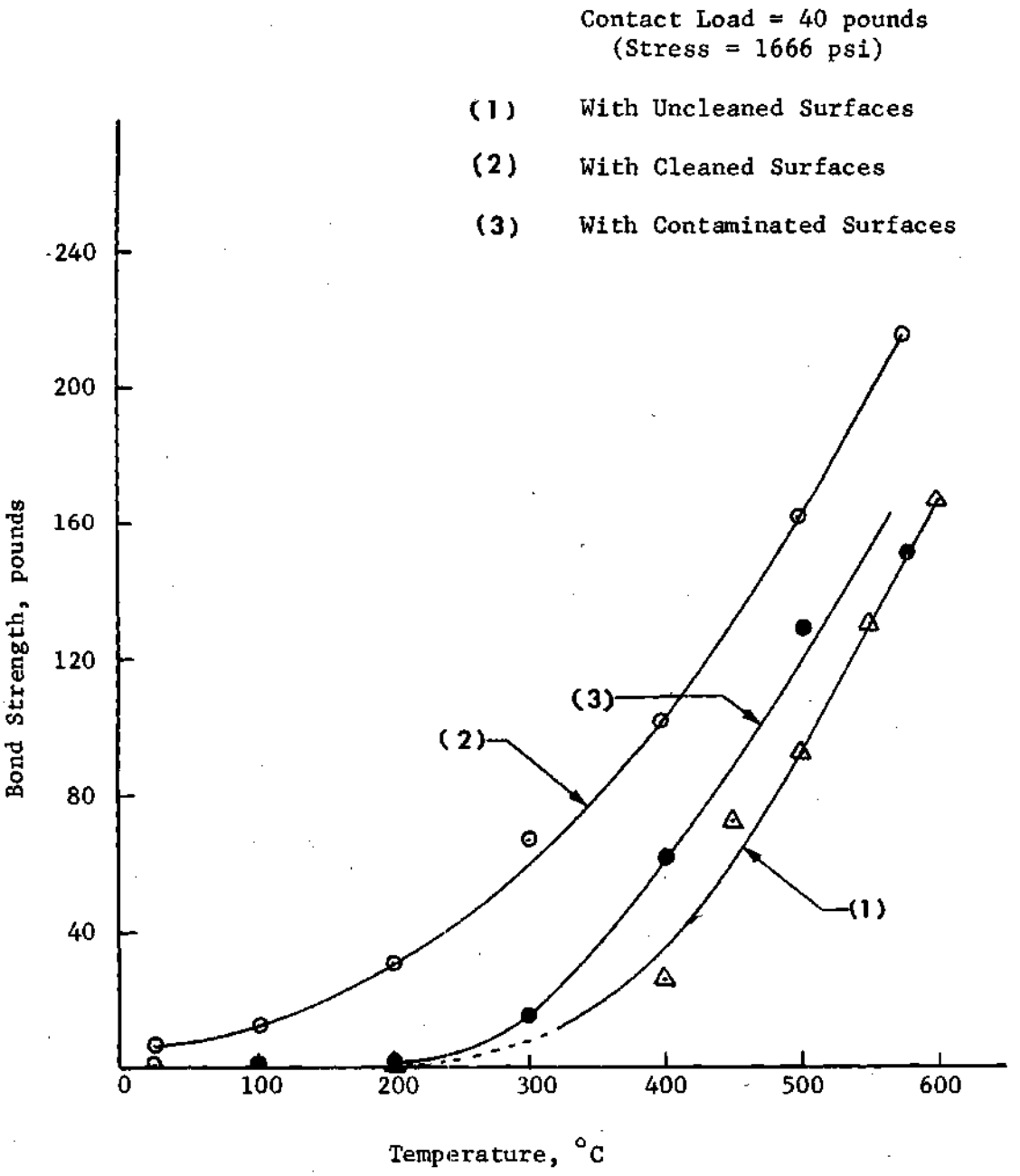


Figure 12. Comparison of Bond Strengths Obtained with Different Surface Cleanliness Conditions

Activation Energy of Bonding Process

Using the recrystallization theory of Johnson and Mehl (42), Ling (33) proposed the following equation for bond strength at any temperature:

$$\sigma = \sigma_o \, e^{-Q/RT} \, t^n \qquad (7)$$

in which σ = dimensionless adhesion (bond strength) at any temperature defined as F/F_c where F = bond strength at any temperature, and F_c = compressive load, σ_o = constant, Q = activation energy of the process, R = gas constant, t = time in contact, and n controls the time dependence of the process. Ling presented data taken for copper couples which were put together for one hour at various temperatures and at a pressure of 10^{-6} torr or higher. No cleaning procedure was used to initially clean the surfaces before they were put into contact. For experiments done at an environmental pressure of 10^{-6} torr, he obtained data that fit equation (7) with Q comparable to the activation energies of bulk self-diffusion in the metals studied. Surprisingly, however, his data varied greatly with environmental pressure. With experiments done at 760 mm Hg, he obtained Q of approximate 4 kcal/g-mole for copper, which is much less than the activation energy of bulk self-diffusion for copper.

To compare the data obtained in this study with the data of Ling (33), Figure 13 is drawn. As is evident from the figure, the relation between 1/T and Log F/F_c is not a straight line for temperatures lower than 200°C. Ling did not present any data below 200°C. For temperatures greater than 200°C, data from tests using clean and contaminated surfaces fall reasonably well on straight lines with different slopes. Assuming

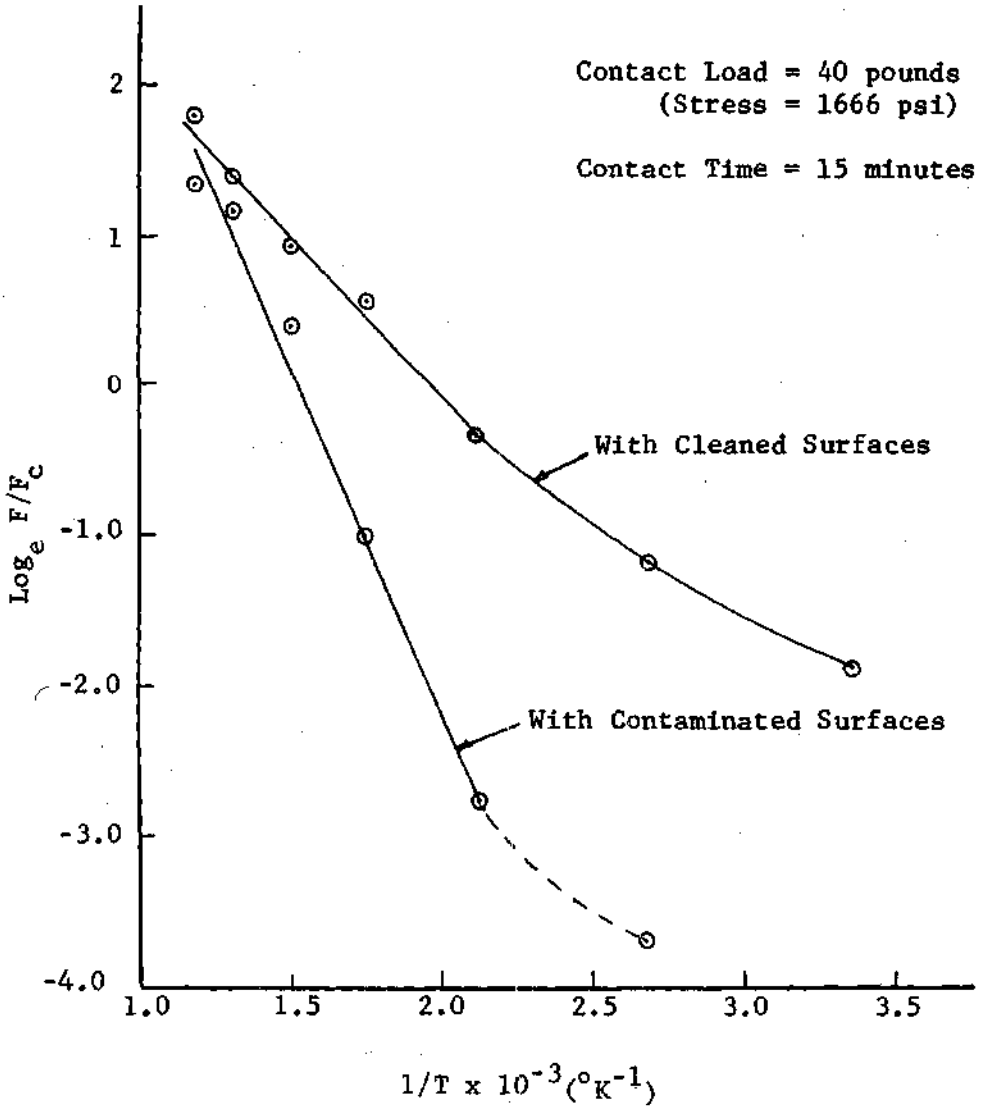


Figure 13. $\text{Log}_e F/F_c$ vs. 1/T

the bonding to be a single thermally activated process, the activation energies as obtained from Figure 13 with clean surfaces is 4 kcal/g-mole and with contaminated surfaces is 9.2 kcal/g-mole.

The activation energy for volume diffusion of copper is reported to be 57.2 kcal/g-mole (43) which is much higher than the activation energy obtained for the bonding process. Ling in his derivation of equation (7) made the basic assumption that the underlying mechanism of bonding is recrystallization or recovery. It therefore seems that for copper the mechanism of bonding is not recrystallization because if it were, the activation energy of the bonding process should have been equal to the activation energy for volume diffusion as suggested by Ling.

Bradford (21) from his study on nickel has suggested that the growth of contact junctions at the joint interface was by surface diffusion. From his results he obtained an activation energy of the process (Q) equal to that for surface diffusion. As discussed by Tylecote (39), the true activation energy of surface diffusion (Q_s) should be about one-fifth that for volume diffusion. If this is true for copper, then the obtained value of activation energy (9.2 kcal/g-mole) for contaminated surfaces almost equals that for surface diffusion. However, the reported value in the literature for copper is higher than one-fifth of its activation energy for volume diffusion. Many workers (44) have reported Q_s for copper to be greater than 40 kcal/g-mole for temperatures greater than 800°C. Bradshaw and co-workers (45), however, found Q_s to be about 20 kcal/g-mole for temperatures between 800°C and 400°C. An important result of their study was that the surface self-diffusion of copper was

found to be dependent on environment. They found that an increase of oxygen pressure increased the surface diffusion. With experiments done in vacuum environment at 1×10^{-7} torr pressure, they obtained a Q_s of 17 kcal/g-mole for surface diffusion. Earlier Menzel (46), using different techniques, had obtained Q_s for copper to be 13 kcal/g-mole for temperatures between 650°C and the melting point in vacuum at 3×10^{-4} torr pressure. Menzel had also noted that changing the environment altered the surface diffusion rate. It should be noted that in the experiments by Bradshaw (45) and Menzel (46), only the environment was controlled. The surfaces were not cleaned and therefore their values for Q_s essentially correspond to uncleaned surfaces. In this study, the value of activation energy for bonding of the uncleaned surfaces was found to be 10 kcal/g-mole, which is of the same order as the value of Q_s given by Menzel (46) and Bradshaw (45). A correlation between activation energy for the bonding process and activation energy for surface diffusion might then exist as suggested by Bradford (21).

The value of activation energy for cleaned surfaces obtained in present investigation is only 4 kcal/g-mole. This value does not compare favorably with the quoted values of activation energy for surface diffusion. The difference could be due to the fact that the quoted values are for uncleaned surfaces. For cleaned surfaces, the activation energy for surface diffusion is not known. It is possible that the activation energy for clean surfaces may be lower than 17 or 13 kcal/g-mole Another possibility,of course, is that the mechanism by which the contact area increases in bonding process is not surface diffusion.

Variation of Bond Strength with Bonding Temperature at Higher Contact Load

Figure 14 shows the results of another similar test series in which the samples were put together with 100 pounds compressive load instead of 40 pounds as done in earlier test series. As the curves show, the joint strength with contaminated surfaces equals the strength obtained with initially cleaned surfaces at bonding temperatures higher than approximately 400°C. This behavior is different from the results obtained in previous test series with compressive load of 40 pounds. The reason for this behavior is that with higher compression greater deformation of the asperities at the interface occurs which may break the contaminant film formed by exposing the bonding surfaces to oxygen. This process of breaking the contaminant film is accelerated at high temperatures as greater deformation at the interface occurs at higher temperatures due to the decrease in yield strength.

Change of Bond Strength with Heating Time

Results of another test series are presented in Figure 15 showing joint strength as a function of heating time at constant load and temperature for cleaned and contaminated surfaces. The surfaces were contaminated as before with 7×10^{-2} torr-sec exposure to oxygen. The curves show that the joint strength increases with increase in contact time but is not a strong function as observed by many other workers (1, 6). The joint strength with cleaned surfaces is always more than that with contaminated surfaces for the range of heating times tested and for the specified conditions of load and temperature. This clearly shows that it is possible to achieve the same bond strength with a lower

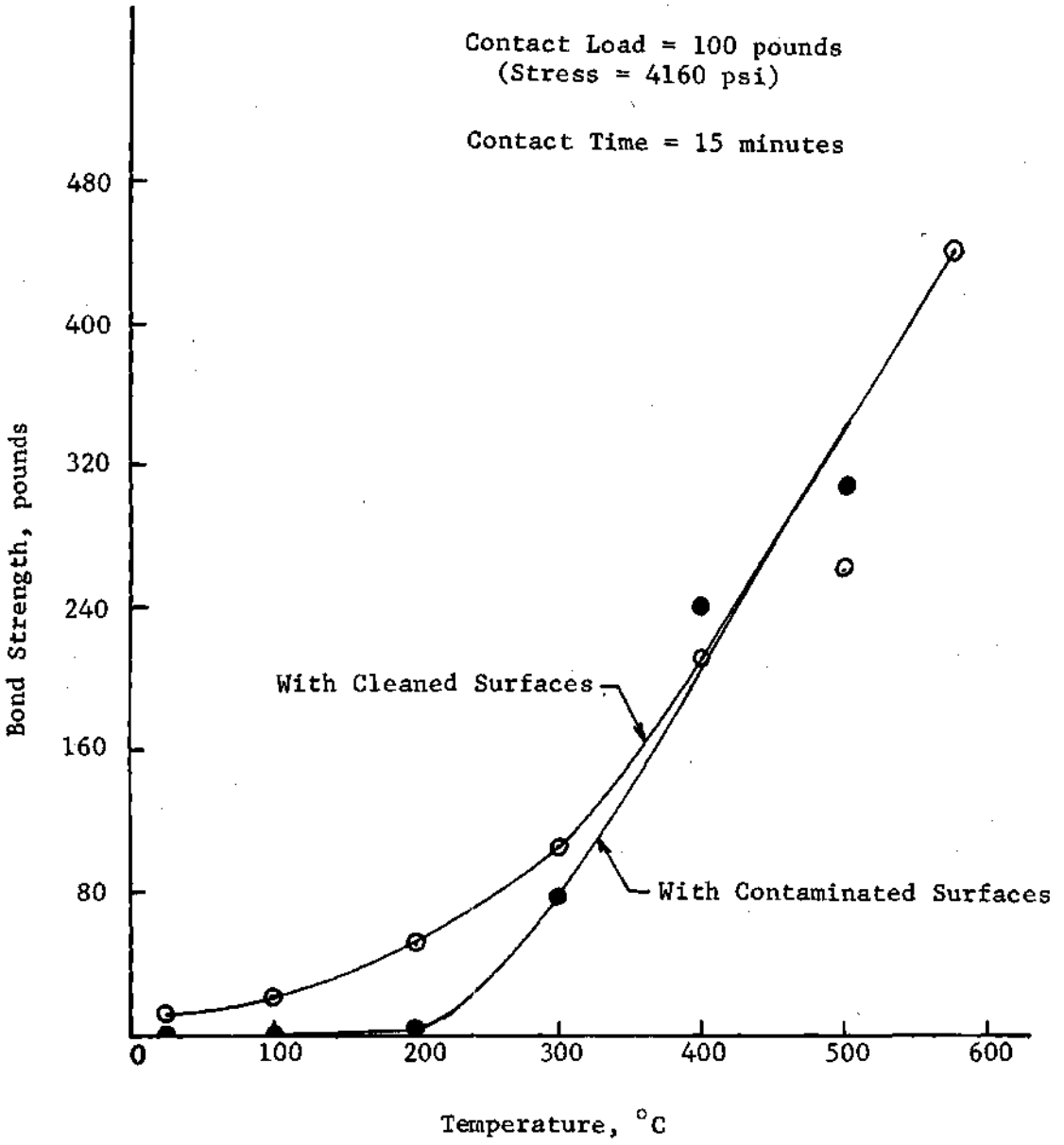


Figure 14. Bond Strength vs. Bonding Temperature for Cleaned and Contaminated Surfaces

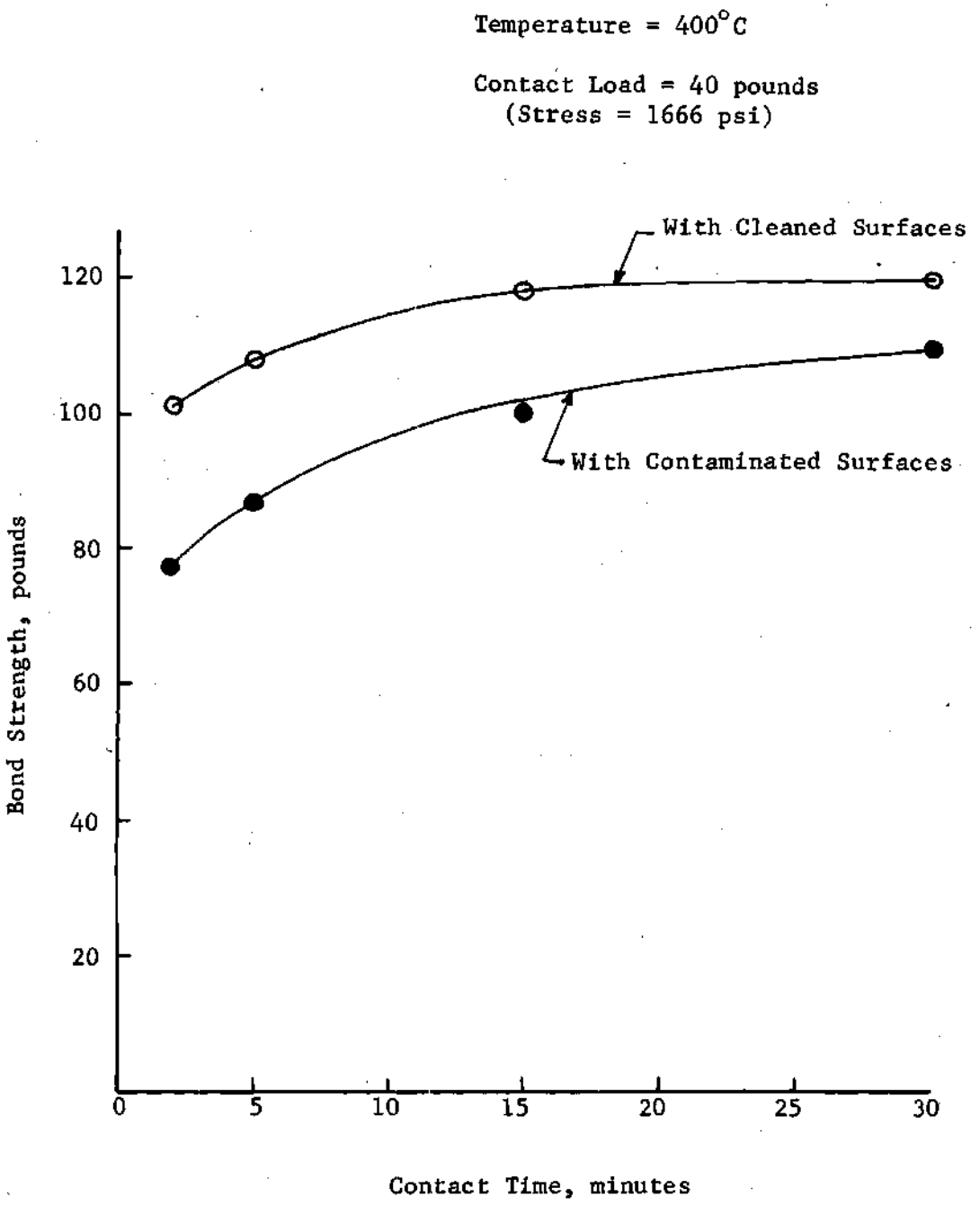


Figure 15. Bond Strength vs. Time
for Cleaned and Contaminated Surfaces

heating time by initially cleaning the surfaces. For instance, with initial cleaning of surfaces a joint strength of 110 pounds (stress = 4560 psi) can be obtained with 6 minutes heating time whereas the time of heating with contaminated surfaces needs to be 30 minutes to obtain the same joint strength. From the curves it can also be seen that difference in joint strengths with cleaned and contaminated surfaces decreases with increased contact time. This is due to the diffusion of contamination into the bulk which is a time-dependent process. As the time of contact is increased, increased diffusion of contamination occurs and reduces the effect of the surface contaminants.

Metallographic Examination

Figure 16 shows the enlarged view of some portions of bonded interfaces with clean and contaminated surfaces. The bonds were made at 400°C. The contact pressure was 100 pounds and the heating time was fifteen minutes. From the photomicrographs it seems that there is no appreciable difference at bonded interfaces for these surface cleanliness conditions. The photomicrographs also show that there is no observable recrystallization across the interface at this magnification. Microhardness testing across the interface showed no change in hardness. A hardness of 58 VHN-200 gms was found over the entire section.

Magnification 550X

Copper to copper

With contaminated surfaces

Load: 100#
Temp: 400°C
Time: 15 minutes

Magnification 550X

Copper to copper

With cleaned surfaces

Load: 100#
Temp: 400°C
Time: 15 minutes

Figure 16. Photomicrograph of Interface

CHAPTER VI

CONCLUSIONS AND RECOMMENDATIONS

The following are the conclusions obtained from this investigation:

1. Copper surfaces can be cleaned by filing in ultrahigh vacuum to the extent that room temperature adhesion coefficients of .15 to .3 can be obtained.

2. For copper, joints made in the temperature range of 25°C to 575°C and with a compressive load of 40 pounds (stress = 1666 psi) are stronger if the contacting surfaces are cleaned prior to bonding. As a corollary it may be said that with cleaned surfaces the same joint strength as obtained with uncleaned surfaces is achieved at a lower bonding temperature for the above specified condition of contact load.

3. The difference in joint strengths obtained with cleaned surfaces and uncleaned surfaces decreases as the bonding temperature is increased for joints made in the temperature range of 25°C to 575°C with a compressive load of 40 pounds (1666 psi).

4. Exposing the cleaned surfaces to oxygen reduces the strength of bond made at 300°C with a compressive load of 40 pounds. With 8.4×10^{-2} torr-sec exposure to oxygen, the strength of bond is reduced from 67 pounds to 20 pounds.

5. Exposing the cleaned surfaces to 7×10^{-2} torr-sec of oxygen does not reduce their joint strength as much as contaminating in air

does for joints made in the temperature range of 25°C to 575°C with a compressive load of 40 pounds.

6. For the case of increase in bond strength with temperature, activation energies of 4 kcal/g-mole and 9.2 kcal/g-mole were obtained with cleaned surfaces and contaminated surfaces, respectively.

7. When the joints are formed with a compressive load of 100 pounds (stress = 4160 psi), stronger bonds are obtained with cleaned surfaces as compared to those obtained with contaminated surfaces for bonding temperatures in the range of 25°C to 400°C. For bonding temperatures between 400°C and 575°C, the joint strengths obtained with the two surface cleanliness conditions are equal.

8. The effect of increasing bonding time is to reduce the difference in joint strengths obtained with cleaned and contaminated surfaces for joints made with a contact load of 40 pounds and at bonding temperature of 400°C.

The following are recommendations for further research based on the results obtained from this investigation:

1. Study the effect of surface cleanliness for other metals to determine if the results of this study for copper can be generalized.

2. Determine the influence of purity of metal on the degree of cleanliness obtained by filing.

3. Determine the effect on solid phase bonding of cold work given to the surfaces by mechanical abrasion.

4. Study the effect of oxygen exposure on bond strength for metals which have no oxygen solubility.

5. Study the mechanism of the growth of the contact area as a function of time.

6. Study the interface for solid phase bonds made at different bonding temperatures by electron microscopy or by some other refined rechnique.

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